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## MEMORANDUM OF INVENTION

To: Patent Department

Date: October 30, 2001Disclosure Number: MD-01-<sup>128</sup>xxx-LSInvention Category: 7Submitted by: Richard R. RoeslerDivision/Dept: LS - Research

## A Title of Invention

Silane terminated polyurethane

## B Business Reason for Invention

Silane terminated polyurethanes (STPs) are useful in the preparation of caulks, sealants and adhesives.

## C Description of Invention

STPs have been prepared with polyether monools and diols using isocyanate functional silanes. When Acclaim diols are used the resulting caulks have excellent tensile strength and elongation. The invention utilizes monosilanes prepared from polyether monools in combination with disilanes to achieve sealants with varying tensile properties. (See attachment for details.)

## D Experimental Work Done on Invention

Date 5/01 through 7/01Notebook Page # 4039373, 40439377,  
4042581-2, 4048214, 4048216

## E Patents, publications, product bulletins, catalogs and other publication which pertain to the invention

WO 9844022—Preparation of polyoxyalkylene monoethers.

WO 9818843—Sealants using silane aspartate

MOI MD-01-66-LS

## F When did the invention first occur to you?

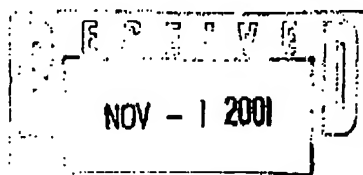
4/26/01

## G To whom at Bayer other than co-inventors did you first communicate your invention?

Various Bayer LS and PU personnel.

## H Has the invention been communicated or sampled to anyone besides Bayer employees?

No.



Witness to Foregoing Description and  
to the Signature(s) of the Inventor(s)

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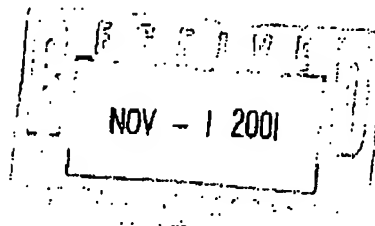
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### Description of Invention

Silane terminated polyurethanes (STPs) have been prepared with polyether diols. When Acclaim polyether diols are used, the resulting caulks have excellent tensile strength and elongation. In addition, the combination of monosilanes prepared from polyether monools and disilanes achieve sealant products with varying tensile properties. Moreover at high levels of monosilane pressure sensitive adhesives can be prepared that retain the excellent tensile properties.

The STPs are prepared by reacting an isocyanatopropyl silane with OH terminated polyether resins.

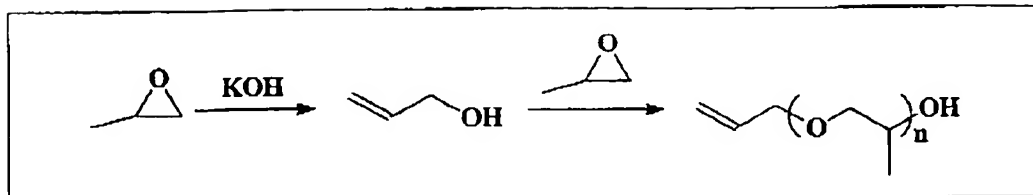
Sealant STPs should have a molecular weight from 4,000 to 20,000. Until now the only way to achieve this molecular weight using polyether prepared with the KOH process was to use 2000 molecular weight diols and prepare isocyanate terminated polyurethane oligomers. The presence of urethane groups causes the products to have high viscosity. The high viscosity requires more plasticizer and less filler pigment, resulting in more expensive sealant products.

The Acclaim diols of this invention have molecular weight starting at 4,000 and rising to 12,000. Preparation of STPs with isocyanatopropyl silane gives products with lower viscosity, and therefore, less plasticizer is required, more pigment can be used leading to lower cost sealants.

Another advantage of using Acclaim polyethers to prepare STPs is control of the mono- and di-ol ratio. The silane functionality of STPs made with Acclaim polyethers can be varied from about two to one. Polyethers prepared with the KOH process results in polyethers with high levels of monool. Two functional STPs cannot be made from KOH based polyethers. (See the following table. MMC is the mixed metal cyanide catalyst.)

Polyol Type	Monool Unsaturation (meq/g)	Functionality	Monool (mole %)
1000 MW diol			
KOH	0.011	1.989	1
MMC	NA	NA	NA
2000 MW diol			
KOH	0.030	1.94	6
MMC	0.005	1.99	1
4000 MW diol			
KOH	0.100	1.67	33
MMC	0.005	1.98	2
8000 MW diol			
KOH	NA	NA	NA
MMC	0.005	1.96	4
12000 MW diol			
KOH	NA	NA	NA
MMC	0.007	1.92	8

The high monool content of the polyether prepared with the KOH process arises because the propylene oxide used to make the polyether isomerizes to allyl alcohol. (See the following Figure.)



Variation of the functionality of the STPs allows different performance properties to be developed to meet the different needs of the user.

An alternative method of making a similar product is by reacting a silane functional amine (e.g., aspartate) with an isocyanate terminated polyol prepolymer. A problem with using an isocyanate prepolymer adduct of silane aspartate is that the urea group increases the viscosity of the resulting resin.

Variation of the functionality of the STPs allows different performance properties to be developed to meet the different needs of the user.

### Examples

#### *Example 1: Preparation of Silane Terminated Polyurethane 1*

A 1 liter round bottom flask was fitted with agitator, nitrogen inlet, condenser, heater and addition funnel. Into the flask was charged 238.5 g (0.041 eq) Impact polyether diol of 12000 molecular/weight (Acclaim 12200, Bayer Corporation), 11.1 g (0.041 eq) 3-isocyanatopropyltrimethoxy silane (Silquest Y-5187, OCI Corporation) and 0.05 g dibutyltin dilaurate. The reaction was heated to 50°C for 4 hours when no NCO remained as determined by an IR spectrum. 1.24 g of vinyl trimethoxysilane was added as moisture scavenger. The viscosity was 4,950 cps @ 25°C. The functionality is two trimethoxysilanes per molecule.

#### *Example 2: Preparation of Silane Terminated Polyurethane 2*

A 1 liter round bottom flask was fitted with agitator, nitrogen inlet, condenser, heater and addition funnel. Into the flask was charged 239.9 g (0.033 eq) Impact polyether monool of 6411 g molecular/weight, 8.91 g (0.033 eq) 3-isocyanatopropyltrimethoxy silane (Silquest Y-5187, OCI Corporation) and 0.05 g dibutyltin dilaurate. The reaction was heated to 50°C for 4 hours when no NCO remained as determined by an IR spectrum. 1.24 g of vinyl trimethoxysilane was added as moisture scavenger. The viscosity was 2,800 cps @ 25°C. The functionality is one trimethoxysilane per molecule.

For reference, but not for inclusion in a patent, the following table gives the Acclaim polyols tradenames for the corresponding polyols used in the above STP #s. [NOTE: the monools do not have a tradename so I have given them the name they would have if they were named according to the established naming criterion.]

STP #	Tradename	Functionality
1	Acclaim 12200	2
2	Acclaim 8200	2
3	Acclaim 4200	2
4	"Acclaim 9100"	1
5	"Acclaim 6100"	1
6	"Acclaim 3100"	1
7	"Acclaim 1100"      Actual tradename: Aktaclear MD21A	1

#### *Formulation of Silane Sealants*

The four STPs were formulated into sealants using the following typical formulation and procedure.

#### *Procedure*

The following is the standard sealant formulation and procedure used to formulate all diol and diol/monool blend experiments. Values given for each formula component are percent by weight of the total formula weight. A high-speed centrifugal mixer was used to mix the formulation components in the steps given below. Each mixing period was one minute in length at a speed of 2200 rpm.

##### *Step 1:*

To a clean dry mixing container was charged the following:

Prepolymer (blend)	37.5
Plasticizer	17.5
Adhesion Promoter	0.8
Catalyst	0.1
Desiccant	0.5

The ingredients were mixed for one minute in length at a speed of 2200 rpm.

##### *Step 2:*

A portion of the filler was added to the mixing container.

Filler	23.6
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The ingredients were mixed for one minute in length at a speed of 2200 rpm.

##### *Step 3:*

The remaining filler was added to the mixing container.

Filler	20.0
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The ingredients were mixed for one minute in length at a speed of 2200 rpm.

**Step 4:**

The side of the mix container was scraped and the ingredients were mixed for one additional minute at a speed of 2200 rpm to incorporate all of the filler into the mixture.

**Step 5:**

The end material was degassed at 50°C and under full vacuum (>28 mm Hg) for one hour. The material was used immediately.

Exxon Jayflex DIDP was used as the plasticizer. CK Witco Silane A-1120 (aminosilane) was used as the adhesion promoter. CK Witco Silane A-171 (vinyltrimethoxysilane) was used as the desiccant. The filler used was Specialty Minerals Ultra P Flex precipitated calcium carbonate (mean particle size of 0.07 microns). The catalyst used was dibutyltin dilaurate.

**Formulation of Silane Sealants**

The two STPs were formulated into sealants using the following typical formulation and procedure.

**Cure and Testing of Silane Sealants**

The sealants from above were cast onto 0.25inch thick polyethylene sheets and cured at standard conditions of 20°C, 50% relative humidity for at least two weeks before testing. Tensile strength, percent elongation and 100% modulus were determined according to ASTM D-412. Die "C" tear strengths were determined according to ASTM D-624. The results are given in the following table.

**Examples 5-8: Tensile Properties for the sealants based on the 12K Diol and 9K Monoool**

Example	Diol-based STP	Monoool-based STP	Diol Monoool Ratio	Die-C Tear (lbs/in)	Ultimate Tensile Strength (psi)	Modulus @ 100% Elongation (psi)	Elongation (%)
5	1	—	—	32	292	188	191
6	1	2	80:20	28	254	203	158
7	1	2	60:40	25	201	179	141
8	1	2	40:60	13	140	187	95

**Extensions of the Art**

The above examples used OH terminated polypropylenes, but other types of mono- and di-functional materials could be used: NH<sub>2</sub> or SH.

The molecular weight for the diol was 12,000 and for the monoool was 7,000 but the usable range for the diol could be 4000 to 12000 and for the monoool could be 1000 to 9000.

The polyether was started with nonyl phenol, but other types of monoool starters could be used, e.g., butanol, allyl alcohol, etc.

$\gamma$ -Isocyanatopropyl trimethoxy silane was used to prepare the silane aspartate, but methyldimethoxysilane or triethoxysilane could be used, as well.